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RPPR Final Report

as of 08-Nov-2018

Agency Code:

Proposal Number: 65441EL

Agreement Number: W911NF-14-1-0356

INVESTIGATOR(S):

Name: Xi-Cheng Zhang
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Principal: Y

Organization: **University of Rochester**

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Country: USA

DUNS Number: 041294109

EIN: 160743209

Report Date: 31-Mar-2018

Date Received: 07-Nov-2018

Final Report for Period Beginning 01-Jul-2014 and Ending 31-Dec-2017

Title: Bright THz Source and Nonlinear Field-Matter Interaction

Begin Performance Period: 01-Jul-2014

End Performance Period: 31-Dec-2017

Report Term: 0-Other

Submitted By: Xi-Cheng Zhang

Email: xi-cheng.zhang@rochester.edu

Phone: (585) 275-0333

Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees:

STEM Participants:

Major Goals: The most important achievement among several others during this ARO grant during the closing year is that we have demonstrated broadband THz wave generation from liquid water. THz wave generations from solid, gas, and plasma have been demonstrated, however, THz wave generation from liquid materials, especially from polar liquid such as water, has not been reported. Einstein gave Einstein relation on Einstein coefficients (emission and absorption) 100 years ago. It says that emission and absorption coefficients are related. For water which has gigantic absorption coefficient in THz range, then it should have big emission capability. We are very happy that we are able to demonstrate it in liquid water. SciLight at American Physics Society and University of Rochester will have a press release soon. The students and postdoctoral were interviewed, photos were taken in the lab.

Our aim is to develop a state-of-the-art, time-resolved bright THz source with at least 10 MV/cm THz peak field strength. This one-order-of-magnitude higher field strength THz source will open the door for many research frontiers in THz field-matter nonlinear interaction. We made significant progress in the following aspects last year: (1) Demonstrate broadband THz generation from liquid water. (2) Terahertz Radiation Enhanced Emission of Fluorescence (REEF) from elongated plasmas and microplasmas in the counter-propagating geometry. (3) Measurement of an extremely large nonlinear refractive index of crystalline ZnSe at terahertz frequencies by a modified Z-scan method. (4) Squeezing the fundamental temperature fluctuations of a high-Q microresonator.

Accomplishments: Papers published in peer reviewed journals (including submitted)

1. Jin Qi, Yiwen E, Kaia Williams, Jianming Dai, X.-C. Zhang, "Observation of Broadband THz Generation from Liquid water", Appl. Phys. Lett, 111, (2017).
2. Xuan Sun, Rui Luo, X.-C. Zhang, Qiang Lin, Squeezing the fundamental temperature fluctuations of a high-Q microresonator, Physical Review A 95, (2017), 023822.
3. Xuan Sun, Hanxiao Liang, Rui Luo, Wei C. Jiang, Xi-Cheng Zhang, and Qiang Lin, "Nonlinear optical oscillation dynamics in high-Q lithium niobate microresonators," Opt. Express 25, 13504-13516 (2017)
4. A.P. Shkurinov, A.S. Sinko, P.M. Solyankin, A.V. Borodin, M.N. Esaulkov, V.V. Annenkov, I.A. Kotelnikov, I.V. Timofeev, X.-C. Zhang, Impact of the dipole contribution on the terahertz emission of air-based plasma induced by tightly focused femtosecond laser pulses, Physical Review E 95, (2017), 043209.
5. Kang Liu, D. G. Papazoglou, A. D. Koulouklidis, S. Tzortzakis, and X.-C. Zhang, "Enhanced terahertz wave emission from air-plasma tailored by abruptly autofocusing beam," Optica 3(6) 605, (2016).
6. L.L. Zhang, T. Wu, H. Zhao, C. Zhang, W. Jin, X.-C. Zhang, Enhanced THz-to-IR emission from gas-surrounded metallic nanostructures by femtosecond laser irradiation, Optics Communications, 381, (2016), 414-417.
7. Y.A. Kapoyko, A.A. Drozdov, S.A. Kozlov, X.C. Zhang, Evolution of few-cycle pulses in nonlinear dispersive

RPPR Final Report as of 08-Nov-2018

media: Velocity of the center of mass and root-mean-square duration, Physical Review A 94 (2016) 9.

8. Fabrizio Buccheri, Kang Liu, X.-C. Zhang, "Terahertz Radiation Enhanced Emission of Fluorescence from Elongated Plasmas and Microplasmas in the Counter-propagating Geometry," Appl. Phys. Lett., submitted.
9. Anton N. Tsytkin, Sergey E. Putilin, Maksim S. Kulya, et al., Measurement of an extremely large nonlinear refractive index of crystalline ZnSe at terahertz frequencies by a modified Z-scan method, Optical Express, submitted.
10. L.L. Zhang, S.J. Zhang, R. Zhang, T. Wu, Y.J. Zhao, C.L. Zhang, and X.-C. Zhang, "Excitation-Wavelength Dependent Terahertz Wave Polarization Control in Laser-Induced Filament," Optica, (2017). Submitted

b. Non-peer-reviewed conference proceeding publications

1. Q. Jin, Y. E. K. Williams, J. Dai, X.-C. Zhang, Observation of Broadband Terahertz Wave Generation from Liquid Water, Nonlinear Optics, Optical Society of America, Waikoloa, Hawaii, 2017, pp. NW3A.1.
2. Y.V. Grachev, X. Liu, A.N. Tsytkin, S.E. Putilin, V.G. Bespalov, S.A. Kozlov, X.C. Zhang, THz sliced broadband continuum for wireless data transfer with CdSe-CdS modulator, 2016 41st International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz), 2016, pp. 1-2.

Training Opportunities: Nothing to Report

Results Dissemination: Conference Proceedings

a. Presentations at meetings, but not published in conference proceedings

1. "From Speculation to Demonstration: THz Wave Emission from Water," Physics Department, Capital Normal University of Beijing, Beijing, China, June 19, 2017.
2. "Let light shine out of darkness" Princeton International School of Math and Sciences, Princeton, NY, May 12, 2017.
3. "THz: Imaging Beyond Light" Open Readings 2017, 60th International Conference for Students of Physics and Natural Sciences, Vilnius, Lithuania, March 15, 2017.
4. "The Institute of Optics, A jewel in the crown," Academy of Opto-Electronics, Chinese Academy of Science, Beijing, China, Dec. 27, 2016.
5. "The Institute of Optics, Micro-Plasma and Extreme THz Science," Jiliang University, Nov. 8, 2016.
6. "Extreme THz Science," Westlake Photonics Symposium, Zhejiang University, Hangzhou, China, Nov. 7, 2016.
7. "Enhanced terahertz wave emission from air-plasma tailored by abruptly autofocusing laser beams," 8th ISUPTW, Chongqing, China, Oct. 11, 2016
8. "The State of The Institute of Optics," New Graduate students town hall meeting, The Institute of Optics, University of Rochester, September 14, 2016.
9. "Vision" Hong Kong University of Science and Technology, Hong Kong, September 6, 2016.
10. "The Institute of Optics: A Jewel in the Crown," Shandong University of Beijing, Beijing, China, July 18, 2016.
11. "The Institute of Optics: A Jewel in the Crown, Bright THz source and Micro-plasma," Capital Normal University of Beijing, Beijing, China, July 2, 2016.
12. "The Institute of Optics: A Jewel in the Crown," Huazhong University of Science and Technology, Wuhan, China, June 28, 2016.
13. X.-C. Zhang, "Bright THz & Nonlinear THz Field-Matter Interaction," ARO Workshop, University of Rochester, Rochester, USA, June 15, 2016.

Honors and Awards: c. Honor and Awards

- 2016-17 X.-C. Zhang, Scientific Advisor, Capital Normal University of Beijing, China
2017 Kang Liu, Rochester Precision Optics Award for Outstanding Graduate Projects
2017 X.-C. Zhang, Australian Academy of Science Selby Fellow, Australia

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Xicheng Zhang

Person Months Worked: 6.00

Funding Support:

RPPR Final Report
as of 08-Nov-2018

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI

Participant: Boyd Robert

Person Months Worked: 3.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

CONFERENCE PAPERS:

Publication Type: Conference Paper or Presentation

Publication Status: 1-Published

Conference Name: 2016 41st International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz)

Date Received: 18-Aug-2017 Conference Date: 25-Sep-2016 Date Published:

Conference Location: Copenhagen, Denmark

Paper Title: THz sliced broadband continuum for wireless data transfer with CdSe-CdS modulator

Authors: Grachev, Y. V., Liu, X., Tsykin, A. N., Putilin, S. E., Bespalov, V. G., Kozlov, S. A., & Zhang, X. C.

Acknowledged Federal Support: **N**

Publication Type: Conference Paper or Presentation

Publication Status: 1-Published

Conference Name: Nonlinear Optics

Date Received: 18-Aug-2017 Conference Date: 17-Jul-2017 Date Published:

Conference Location: Waikoloa, Hawaii

Paper Title: Observation of Broadband Terahertz Wave Generation from Liquid Water

Authors: Jin, Qi, E, Yiwen, Williams, Kaia, Dai, Jianming, & Zhang, Xi-Cheng.

Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation

Publication Status: 1-Published

Conference Name: Frontiers in Optics

Date Received: 18-Aug-2017 Conference Date: 17-Oct-2016 Date Published: 21-Oct-2016

Conference Location: Rochester, New York

Paper Title: Enhancing THz radiation from two-color laser-induced air-plasma by using abruptly autofocusing beams

Authors: Liu, Kang, Koulouklidis, Anastasios D, PAPAOGLOU, DIMITRIOS G, Tzortzakis, Stelios, & Zhang, Xi-

Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation

Publication Status: 1-Published

Conference Name: 2015 40th International Conference on Infrared, Millimeter and Terahertz Waves

Date Received: 20-Aug-2017 Conference Date: 23-Aug-2015 Date Published:

Conference Location: Hong Kong

Paper Title: Terahertz wave emission from dual color laser-induced microplasma

Authors: F. Buccheri, X.-C. Zhang

Acknowledged Federal Support: **Y**

RPPR Final Report
as of 08-Nov-2018

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: 2015 40th International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz)
Date Received: 20-Aug-2017 Conference Date: 23-Aug-2015 Date Published:
Conference Location: Hong Kong, China
Paper Title: Study of THz emission from ring-Airy beam induced plasma
Authors: Kang Liu; D. G. Papazoglou; A. D. Koulouklidis; S. Tzortzakis; X. -C. Zhang
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: 2014 39th International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz)
Date Received: 21-Aug-2017 Conference Date: 14-Sep-2014 Date Published:
Conference Location: Tucson, AZ, USA
Paper Title: Broadband terahertz wave emission from thin metal films excited by two-color laser fields
Authors: Jianming, Dai; X.-C. Zhang
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: 2014 39th International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz)
Date Received: 21-Aug-2017 Conference Date: 14-Sep-2014 Date Published:
Conference Location: Tucson, AZ, USA
Paper Title: Terahertz wave emission from laser-induced micro-plasma
Authors: F. Buccheri, and X.-C. Zhang
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
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Date Received: 21-Aug-2017 Conference Date: 14-Sep-2014 Date Published:
Conference Location: Tucson, AZ, USA
Paper Title: THz wave generation from cesium vapor
Authors: Xuan Sun ; X.-C. Zhang
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: 2014 39th International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz)
Date Received: 21-Aug-2017 Conference Date: 14-Sep-2014 Date Published:
Conference Location: Tucson, AZ, USA
Paper Title: Resonant field enhancement in periodically arranged microslits for non-linear terahertz spectroscopy
Authors: Pernille Klarskov¹, Krzysztof Iwaszczuk¹, Maksim Zalkovskij¹, Radu Malureanu¹, Jianming Dai², X.-C.
Acknowledged Federal Support: **N**

DISSERTATIONS:

RPPR Final Report
as of 08-Nov-2018

Publication Type: Thesis or Dissertation

Institution: Optics

Date Received: 20-Aug-2017

Completion Date: 5/15/17 8:58PM

Title: Terahertz Photonics Based on Resonance-Enhanced Nonlinear Effects

Authors: Xuan, Sun

Acknowledged Federal Support: **Y**

Publication Type: Thesis or Dissertation

Institution: Optics

Date Received: 20-Aug-2017

Completion Date: 1/6/17 5:00AM

Title: Generation and Detection of Pulsed Terahertz Waves with Laser Induced Microplasmas

Authors: Fabrizio Buccheri

Acknowledged Federal Support: **Y**

Publication Type: Thesis or Dissertation

Institution: Optics

Date Received: 20-Aug-2017

Completion Date: 5/20/17 4:00AM

Title: Electromagnetic Wave Generation from Liquid Water

Authors: Qi, Jin

Acknowledged Federal Support: **Y**



ARO Final Report 2017

X.-C. Zhang, University of Rochester



Bright THz source and nonlinear field-matter interaction

Final Progress Report

Report Period: 08/01/2016--07/31/2017

Grant Contract #: W911NF-14-1-0356

Co-Principal Investigators: **X.-C. Zhang** and **Robert Boyd**

The Institute of Optics

University of Rochester

275 Hutchison Road, Rochester, New York 14627

zhangxc@rochester.edu

Authors: Yiwen E, Kang Liu, Robert Boyd, and Xi-Cheng Zhang

University of Rochester

Submitted: Dr. Joe Qiu, ARO

Abstract: The most important achievement among several others during this ARO grant during the closing year is that we have demonstrated broadband THz wave generation from liquid water. THz wave generations from solid, gas, and plasma have been demonstrated, however, THz wave generation from liquid materials, especially from polar liquid such as water, has not been reported. Einstein gave Einstein relation on Einstein coefficients (emission and absorption) 100 years ago. It says that emission and absorption coefficients are related. For water which has gigantic absorption coefficient in THz range, then it should have big emission capability. We are very happy that we are able to demonstrate it in liquid water. SciLight at American Physics Society and University of Rochester will have a press release soon. The students and postdoctoral were interviewed, photos were taken in the lab.

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1. Publications, Presentation and Honors

Papers published in peer reviewed journals (including submitted)

1. Jin Qi, Yiwen E, Kaia Williams, Jianming Dai, X.-C. Zhang, "Observation of Broadband THz Generation from Liquid water", Appl. Phys. Lett, 111, (2017).
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Conference Proceedings

a. Presentations at meetings, but not published in conference proceedings

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2. "Let light shine out of darkness" Princeton International School of Math and Sciences, Princeton, NY, May 12, 2017.
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7. "Enhanced terahertz wave emission from air-plasma tailored by abruptly autofocusing laser beams," 8th ISUPTW, Chongqing, China, Oct. 11, 2016
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12. "The Institute of Optics: A Jewel in the Crown," Huazhong University of Science and Technology, Wuhan, China, June 28, 2016.
13. X.-C. Zhang, "Bright THz & Nonlinear THz Field-Matter Interaction," ARO Workshop, University of Rochester, Rochester, USA, June 15, 2016.

b. Non-peer-reviewed conference proceeding publications

1. Q. Jin, Y. E. K. Williams, J. Dai, X.-C. Zhang, Observation of Broadband Terahertz Wave Generation from Liquid Water, Nonlinear Optics, Optical Society of America, Waikoloa, Hawaii, 2017, pp. NW3A.1.
2. Y.V. Grachev, X. Liu, A.N. Tsyarkin, S.E. Putilin, V.G. Bespalov, S.A. Kozlov, X.C. Zhang, THz sliced broadband continuum for wireless data transfer with CdSe-CdS modulator, 2016 41st International Conference on Infrared, Millimeter, and Terahertz waves (IRMMW-THz), 2016, pp. 1-2.

c. Honor and Awards

2016-17	X.-C. Zhang, Scientific Advisor, Capital Normal University of Beijing, China
2017	Kang Liu, Rochester Precision Optics Award for Outstanding Graduate Projects
2017	X.-C. Zhang, Australian Academy of Science Selby Fellow, Australia

2. Personnel Metrics

Please complete the below tables, providing the information for this reporting period only. Add rows as needed.

1. Graduate Students

Name	Discipline	Percent Supported
Kang Liu	Optics	100%
Mahmudur Siddiqui	Optics	100%
Fabrizio Buccheri	Optics	20%
Kareem Garriga	Optics	20%
Xuan Sun	Optics	20%
Qi Jin	Optics	20%

2. Post Doctorates

Name	Percent
------	---------

	Supported
Anton Koroliov	20%
Dimitar Valchev	0%
Rui Wang	0%
Yiwen E	0%

3. Faculty

Name	National Academy Member	Percent Supported
Xi-Cheng Zhang		5%
Robert Boyd		0%
Jianming Dai		0%

4. Undergraduate Students

Name	Discipline	Percent Supported
Kaia Williams	Optics	0%

5. Other Staff

Name	Percent Supported
None	

3. Graduating Undergraduate Metrics

Please provide a count for each category below for **Graduating Undergraduates** that were funded by this project and graduated during this reporting period.

Category	Number of Undergraduates
Number who graduated during this period	2
Number who graduated during this period with a degree in science, mathematics, engineering or technology fields	2
Number who graduated during this period and will continue to pursue a graduate of Ph.D. in science, math, engineering, or technology fields	0
Number who achieved a 3.5 GPA to 4.0 (4.0 max scale)	0
Number funded by a DoD Funded Center of Excellence grant for Education, Research and Engineering	0
Number who intend to work for the Department of Defense	0
Number who will receive scholarships or fellowships for further studies in science, math, engineering, or technology fields	0

4. Masters Degrees Awarded

Please complete the following table, adding rows as necessary.

Name	Discipline
N/A	

5. Ph.Ds Awarded

Please complete the following table, adding rows as necessary.

Name	Discipline
Xuan Sun	Optics
Fabrizio Buccheri	Optics

6. Technology Transfer

No technology transfer for this reporting period.

None

7. Scientific Progress and Accomplishments

(1) Observation of broadband THz generation in liquid water

We experimentally demonstrate the generation of broadband THz waves from liquid water excited by femtosecond laser pulses. A typical THz time-domain spectroscopy (THz-TDS) is applied to generate and detect our THz signals from liquid water. An amplifier laser with 800 nm wavelength, 1 kHz repetition rate and 50 fs pulse duration is used. A gravity-driven wire-guided free-flowing water film acts as the emitter for the THz field. The thickness of the water film can be adjusted by throttling the flow rate of the water to the waveguide. The thickness is measured and calibrated using an optical intensity autocorrelation system and is 170 μm . The laser beam is focused into the water film using a 1-inch focal length parabolic mirror.

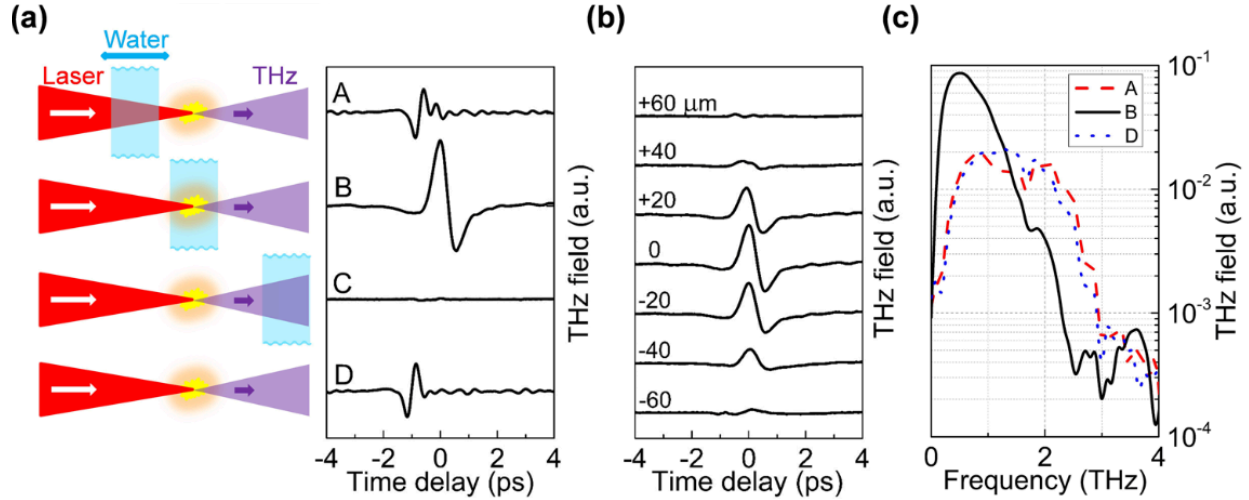


FIG. 1. Measurements of the THz fields when the water film is translated along the direction of laser propagation. (a) THz waveforms are plotted from curves A–C when the water film is before, near, and after the focus, respectively. (b) THz waveforms when the water film is moved near the focal point. (c) Comparison between the THz field from water and that from air plasma in the frequency domain.

By scanning the water film along the optical axis, THz radiation from different sources can be clearly differentiated. The timing distinctions in the waveforms in Fig. 1(a) are of different generation sources. A time delay is observed from the THz waveform from liquid water compared with other generations. Figure 1(b) shows the measurements of THz waveforms as the water film is tracked along the direction of laser propagation marking a relative position across $-60 \mu\text{m}$ to $+60 \mu\text{m}$. The measurement shows that the emitted THz waves are significantly sensitive to the relative position between the water film and the focus. A comparison of the THz waveforms from liquid water and air plasma is shown in Fig. 1(a). In this measurement, the THz field from the water film is 1.8 times stronger than that from the air plasma. The corresponding comparison in the frequency domain is shown in Fig. 1(c). The measured bandwidth can be limited by the stretch of the probe laser pulses. The measured THz radiation from the water has more low-frequency and less high-frequency components.

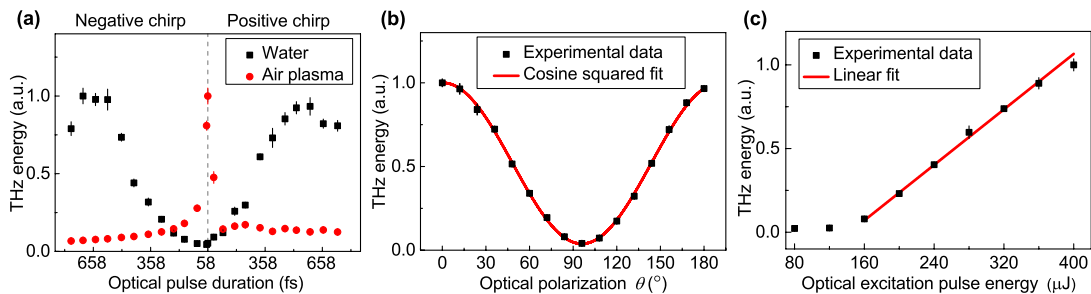


FIG. 2. (a) Normalized THz energy from liquid water and air plasma with different pulse durations of the laser beam. (b) The energy of P-polarized THz field from liquid water with different linearly optical polarization. (c) Normalized THz energy from liquid water as a function of incident optical pulse energy.

Compared with THz radiation generated from air plasma, the THz radiation from liquid water has a distinct response to various optical pulse durations and shows linear energy dependence upon incident laser pulses. Fig. 2(a) shows normalized THz energy from water and air plasma versus various optical pulse durations. The optical pulse duration is at its minimum of 58 fs when no chirp is applied. It can be observed that unlike the THz radiation from air plasma, where the signal is maximized at a minimum pulse duration with no additional chirp, liquid water generates a maximum field at longer pulse durations. With a longer pulse duration, cascade ionization dominates the process leading an exponential increase

in the number of electrons. The THz radiation from liquid water may benefit from higher density of electrons in the water. Fig. 2(b) is shown that strong THz radiation is achieved with a p-polarized (0° and 180°) optical beam, while an s-polarized (90°) optical beam offers sparse contribution. This result goes against the case of single color air plasma THz generation, in which the ponderomotive force is dominantly involved. It is well known that the THz radiation from air plasma with single color optical excitation does not depend upon the polarization of the optical beam, which means the THz radiation energy will keep constant with various optical polarizations. Furthermore, a linear energy dependence observed in Fig. 2(c) is different from the quadratic relation of the single-color air plasma THz generation.

Our first paper which submitted to APL will be published in August. Additionally, when I submitted the abstract to IRMMW-THz annual meeting, within 2 minutes, I got a reply from the conference chair to congratulate our achievement, and 5 congratulations from 4 countries in two days (they might be the paper evaluators). Now we have received 7 plenary and invited talks at international conferences. I am very excited about it.

(2) Terahertz Radiation Enhanced Emission of Fluorescence from Elongated Plasmas and Microplasmas in the Counter-propagating Geometry

Remote sensing is one of the major challenges for Terahertz (THz) radiation applications, due to the THz wave attenuation by the atmosphere water vapor during its propagation. THz-Radiation-Enhanced-Emission-of-Fluorescence (REEF) is a THz air-photonics technique that has the potential to bypass this issue, by having the sought-after THz spectral fingerprints carried from the target to the operator by ultraviolet light, which experiences low absorption in the atmosphere. This technique has been previously demonstrated when the THz radiation and the laser excitation are focused collinearly, namely the co-propagating geometry. However, the co-propagating geometry is not a favorable configuration for practical stand-off detection. Therefore, further exploration on alternative sensing geometries is still required. We studied the interaction of broadband THz radiation with plasmas induced by a counter-propagating laser beam, which is a more desirable geometry for remote sensing. plasma interaction and highlight the potential of THz-REEF technique in the plasma diagnostic applications.

The setup implements the standard pump-probe THz time-domain spectroscopy technique with the relative time delay Δt between the pump and the probe pulses controllable through a motorized linear stage. The laser employed was Spectra Physics Hurricane (800 nm center wavelength, 100 fs pulse duration, 0.8 mJ pulse energy, 1 kHz repetition rate). Intense single cycle THz pulses with peak field of ~ 90 kV/cm were obtained via optical rectification in LiNbO₃ with tilted pulse front geometry. The THz radiation was collected and refocused onto the plasma formed by focusing the optical beam. The plasma fluorescence is imaged into the input slit of a grating monochromator and measured with a photomultiplier tube (PMT) placed at its output slit, or into a gated iCCD camera (PIMAX3 Princeton Instruments). As a reference, the THz time-domain waveform was also measured by electro-optic (EO) sampling. Through flip mirrors, the setup could be easily switched between the co-propagating and counter-propagating geometries, both of which are shown in FIG. 3(a).

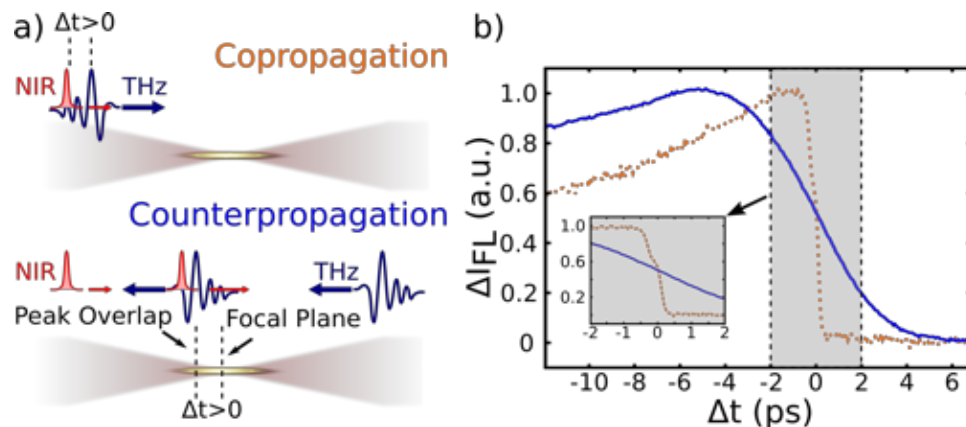


FIG 3. (a) Depiction of the two interaction geometries. (b) Plasma fluorescence intensity enhancement as a function of Δt in co-propagating (orange, dashed) and counter-propagating (blue, solid) geometries measured by PMT.

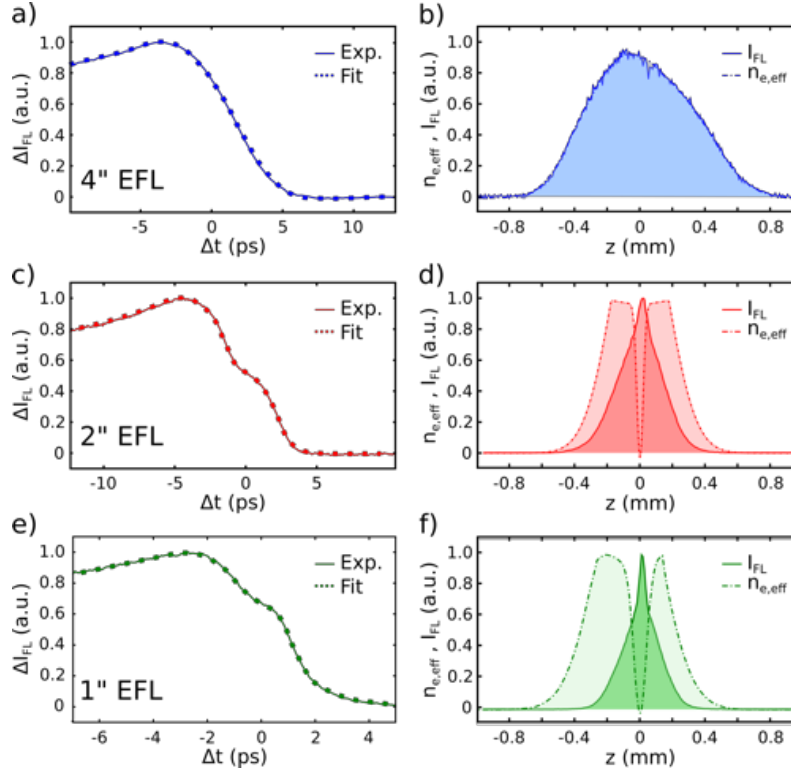


FIG. 4. Experimental (solid line) and numerical fitted (dashed line) plasma fluorescence intensity enhancement as a function of Δt in counter-propagating geometry for the following focusing conditions (a) 4" EFL PC lens; (c) 2" EFL PC lens; (e) 1" EFL PC lens; in figures (b), (d) and (f) the solid lines represent the integration of the plasma fluorescence intensity along the radial dimension as measured with the iCCD camera, whereas the point-and-dash lines are the numerically evaluated plasma effective electron densities producing the dashed curves plotted in (a), (c) and (e).

The comparison between the REEF traces measured by the PMT in both configurations is shown in FIG 3(b). Those plots depict the absolute fluorescence enhancement, obtained by subtracting the background fluorescence, i.e., the fluorescence intensity in absence of THz radiation, from the signal measured with the THz radiation, as a function of Δt . In both cases, the fluorescence signal starts increasing when the THz pulse begins to overlap temporally with the optical pulse at the plasma location. In both configurations the fluorescence enhancement magnitude, defined as the difference between the maximum value of the fluorescence intensity and the fluorescence background, is similar. However, the co-propagating trace (orange, dashed) shows a rapid increase of fluorescence with a timescale comparable to the THz pulse time duration, while the counter-propagating one exhibits a gentler slope with rising time close to 10 ps. To explore this phenomenon in more details, we shortened the dimension of the plasma by changing the focal length of the focusing optics to the point of creating microplasmas.

The time-resolved REEF traces measured by PMT of the plasmas obtained with three different focusing elements, 4", 2" and 1" EFL (Effective Focal Length) in counter-propagation geometry are plotted in FIG. 4(a), (c) (e) and their corresponding numerical fitted plasma fluorescence intensity along the radial dimension (FIG. 4(b), (d), (f)). In the case of elongated plasma (4" EFL), the effective electron density well matches with the fluorescence profile measured with the iCCD camera, therefore indicating that all the plasma volume contributes to the REEF interaction. However, the computed effective index and the measured fluorescence profiles differ dramatically in the cases of the smaller plasmas. Those curves

suggest that the denser part of the plasma has very little contribution to the fluorescence enhancement, whereas the biggest contribution is due to the interaction of the THz wave with the outer region of the plasma, where the electron density is lower. This could be qualitatively explained by two physics phenomena: (i) the volumes with highest electron densities are the ones presenting the highest degree of ionization of the air molecules. It is therefore plausible that the contribution of those volumes to the THz-REEF signal is very small, as the density of electronic states right below the continuum (~ 100 meV), which are the one contributing to the THz induced fluorescence enhancement⁹, is greatly reduced for highly ionized molecules. (ii) As the fluorescence intensity enhancement peculiar to the REEF detection mechanism is the result of the interaction of the THz radiation with a formed plasma⁹, and the estimated characteristic ionization times in our experiment are less than 200 fs, one should also consider the skin effect. For values of electron densities higher than 10^{16} cm⁻³, which are achieved in the microplasmas generated in our experiments, the plasma frequency becomes greater than 1 THz. Frequencies below that value are not allowed to propagate through the plasma, but they decay exponentially within a lengthspace defined by the skin depth of the plasma at the specified frequency. The skin depth gets smaller as the electron density increases. For the estimated electron densities of the experiment and the peak frequency of the input THz pulse (0.7 THz), the skin depth in the densest volume is as low as 5 μ m. Therefore, the densest part of the plasma is screened from the incoming THz pulse and do not contribute to the THz-REEF signal.

(3) Measurement of an extremely large nonlinear refractive index of crystalline ZnSe at terahertz frequencies by a modified Z-scan method

We studied the measurement of the intensity-dependent refractive index n_2 of crystalline ZnSe at THz frequencies. We use a modified Z-scan method with a strong THz beam of maximum intensity 0.8×10^9 W/cm². We measure an n_2 value of the order of 2.5×10^{-11} cm²/W, which is significantly larger than the n_2 value of ZnSe in the near infrared and is also much larger than the n_2 value of typical dielectric materials at optical frequencies. Our results confirm a recent theoretical prediction that the ionic vibrational contribution to the third-order susceptibility makes renders THz nonlinearities much larger than typical optical-frequency nonlinearities.

We measured the amplitude of the THz radiation by means of the electro-optic effect in a ZnTe crystal that is place between the crossed polarizers. When we removed the block of the THz radiation, and using a delay line we found the maximum intensity of the femtosecond probe radiation (corresponding to the maximum of the intensity of the THz pulse) which passed through crossed polarizers. We fixed the position of the delay line of the probe femtosecond pulse relative to the THz pulse at this maximum amplitude. Then, as in the conventional Z-scan method, we translated the ZnSe crystal along the z axis. In distinction from the classical Z-scan method, in which one measures the change of energy in the central part of the beam, we measure the change of intensity of the probe beam in the region of maximum amplitude of the THz pulse. We measure the intensity using a CCD camera having 24-bit intensity resolution. We sum the signal from an array of 20 x 20 pixels in the region of maximum intensity and take this value to represent the amplitude modulus of the THz signal for this particular location in z.

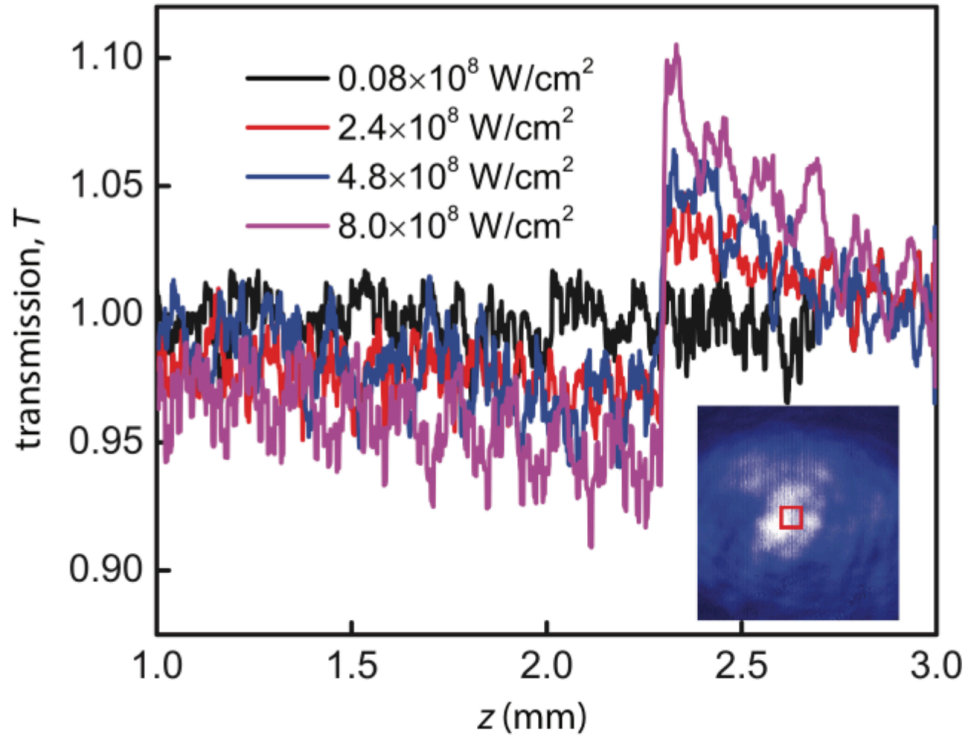


FIG. 5. Z-scan curves for various values of the peak intensity of the THz radiation. T is the transmission through the closed aperture. The red square box in the image of the beam shows the region on the beam axis where the measurement of pulse energy was made.

Z-scan traces for different intensities of the THz radiation are presented in FIG. 5. The vertical axis gives the fractional change in collected power of the probe fs radiation after passing through the crossed polarizers surrounding the electro-optic crystal. Translation of the ZnSe crystal along the z -axis leads to a variation of the total collected power. This sort of variation is characteristic of Z-scan measurements. We use the standard formulas for Z-scan measurements to estimate the nonlinear refractive index (defined by $n = n_0 + n_2 I$). Measurements of the nonlinear refractive index were carried out for various intensities of the THz radiation. From our measurements, we find that $n_2 = 2.5 \pm 1.0 \times 10^{-11} \text{ cm}^2/\text{W}$.

(4) Squeezing the fundamental temperature fluctuations of a high-Q microresonator

Temperature fluctuations of an optical resonator underlie a fundamental limit of its cavity stability. We verified that the fundamental temperature fluctuations of a high-Q microresonator can be suppressed remarkably by pure optical means without cooling the device temperature. An optical wave launched into the cavity is able to produce strong photothermal backaction which dramatically suppresses the spectral intensity of temperature fluctuations and squeezes its overall level by orders of magnitude. The proposed photothermal temperature squeezing is expected to significantly improve the stability of optical resonances, with potentially profound impact on broad applications of high-Q cavities in sensing, metrology, and nonlinear and quantum optics.

The underlying physical mechanism is the photothermal backaction between the device temperature and the intracavity optical energy, as schematically shown in Fig. 6. For an optical wave launched into the cavity, a small fraction of the energy would be absorbed by the device material and converted into heat. The temperature fluctuations of the device modulate the resonance frequency of the cavity, which in turn perturbs the intracavity energy. Consequently, the magnitude of photothermal heating changes accordingly which eventually back-acts onto the device temperature itself. Such a photothermal backaction mechanism underlies various thermo-optic nonlinear phenomena. However, its intriguing

interaction with the fundamental thermo-optic noises of a high-Q resonator has been neglected in the past explorations. Photothermal backaction exhibits very profound impact on the characteristics of the fundamental temperature fluctuations, squeezing its amplitude and the resulting thermorefractive noises by orders of magnitude.

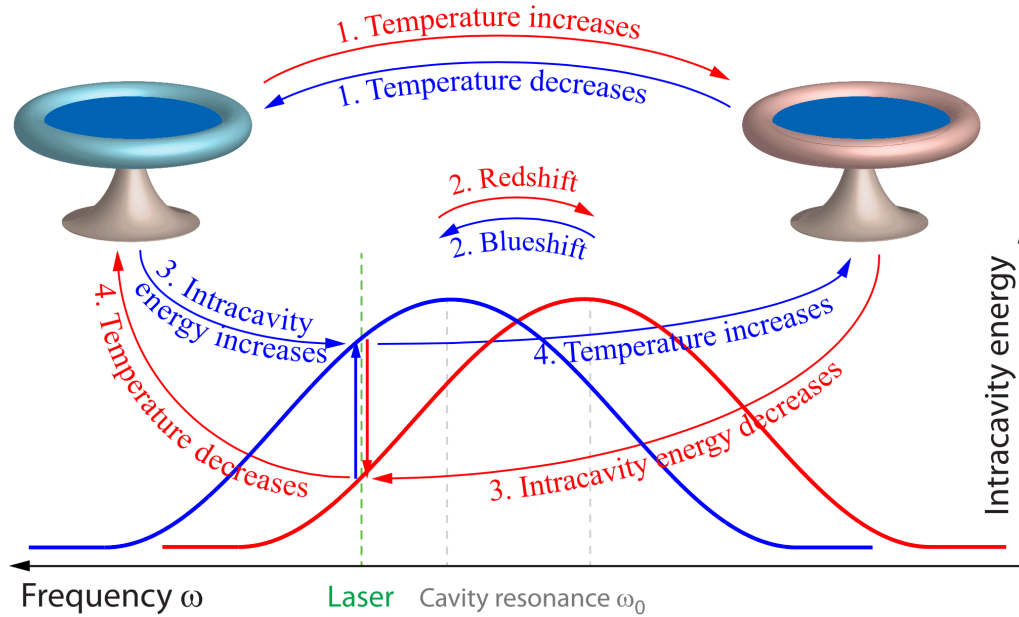


FIG. 6. Schematic of photo thermal backaction to squeeze the temperature fluctuation of a device (shown as a microtoroid). A variation of device temperature (1) shifts the resonance frequency of the cavity (2) (assuming $dn/dT > 0$), resulting in a perturbation to dT the intracavity optical energy (3). As a result, the magnitude of photothermal heating changes, which in turn leads to a backaction on the device temperature (4).

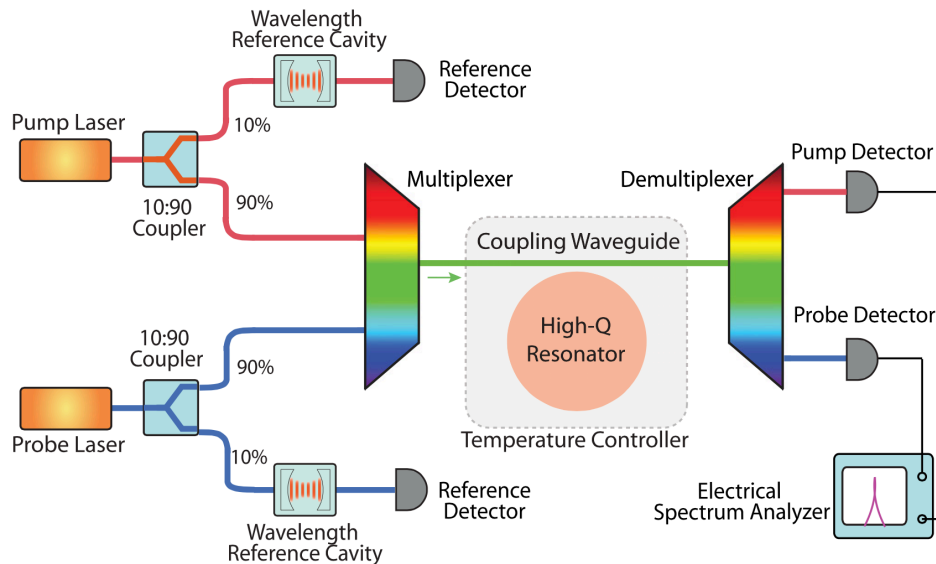


FIG. 7. Schematic of proposed experimental setup for characterizing the temperature squeezing.

We propose an experimental scheme, as shown in Fig. 7, to measure the temperature squeezing. A strong pump laser is launched into a cavity mode of the high-Q resonator to produce temperature squeezing. At the same time, a weak probe laser is launched into a separate cavity mode (within the same mode family of the pump mode) to detect the induced temperature squeezing. Both lasers are locked to stabilized reference cavities (or wavelength references) to avoid potential wavelength drifts. The environmental temperature of the microresonator is stabilized to avoid potential temperature drift. The power spectra of the pump and probe waves output from the resonator can be measured by optical detectors and an electrical spectrum analyzer. Such a testing scheme would provide detailed characterization of the temperature squeezing effect. In practice, one potential interference might come from the optomechanical oscillation excited by the intense pump wave. As this effect depends sensitively on the quality factor of the mechanical mode, it can be easily quenched by introducing mechanical damping to the device.